Non-adiabatic preparation of spin crystals with ultracold polar molecules

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We study the growth dynamics of ordered structures of strongly interacting polar molecules in optical lattices. Using dipole blockade of microwave excitations, we map the system onto an interacting spin-1/2 model possessing ground states with crystalline order, and describe a way to prepare these states by non-adiabatically driving the transitions between molecular rotational levels. The proposed technique bypasses the need to cross a phase transition and allows for the creation of ordered domains of considerably larger size compared to approaches relying on adiabatic preparation.

Long-range dipolar interactions enable the creation of novel states of matter with ultracold quantum gases [1]. A prominent example is the predicted formation of dipolar crystals with tunable interaction parameters [2–4]. However, the preparation of such strongly interacting phases starting from a weakly interacting quantum gas is known to be very challenging, as it involves crossing a phase transition, where the energy gap vanishes in the thermodynamic limit [5]. In this Letter we show that for ultracold polar molecules in an optical lattice, this problem can be overcome by non-adiabatic driving of rotational transitions.

Our approach builds on recent experimental advances in the coherent creation and control of ultracold polar alkali metal dimers, such as KRb [6] and LiCs [7]. The experiments have demonstrated that ultracold polar molecules can now be produced in the rovibrational ground state, transferred to any hyperfine sublevel [8], and trapped in a periodic potential of an optical lattice [9]. Ultracold molecules trapped on an optical lattice have previously been proposed as promising candidates for quantum computation [10] and quantum simulation of spin-lattice models [1, 11, 12]. Here, we employ dipole blockade of microwave excitations in the context of an effective spin-1/2 model with ultracold molecules on an optical lattice. The ground state phase diagram of such model is dominated by a series of commensurate phases, in which one of the spin states exhibits crystalline order. We show that these phases can be prepared by a short sequence of microwave pulses that nucleate the ordered domain, followed by continuous microwave driving that propagates the domain boundary. We provide an effective model that describes the dynamics under the continuous driving and demonstrate that it leads to an efficient growth of the ordered domains. Finally, we analyze the imperfections and the required experimental parameters, and demonstrate that structures consisting of $\gtrsim 1000$ spins can be grown.

To explore these ideas, we consider a one-dimensional array of $^1\Sigma$ molecules with a dipole moment d prepared in a Mott insulator state in an optical lattice with the period a, tilted by the angle θ with respect to the laboratory z axis, as shown in Fig. 1 (a). The lattice is filled with one molecule per site and the trapping potential is strong enough to prevent tunneling between the sites and molecular reactions [9]. The rotational states of a $^1\Sigma$ diatomic molecule, $|J, M\rangle$, are labeled by the angular momentum J and its projection on the laboratory z-axis, M. The energy of the rotational states is given by $E_{\rm rot} = BJ(J+1)$, where B is the rotational constant. We assume that the molecules are initially prepared in the rotational ground $J = 0$ state.

The states $|J, M\rangle$ can be coupled by a microwave field, producing linear combinations of rotational states with different parity. In particular, the rotational states $|0, 0\rangle$ and $|1, 0\rangle$ can be coupled by near-resonant linearly polarized microwave field with Rabi frequency Ω_c and detuning Δ_c to produce two field-dressed states separated by $\sim \Omega_c$. If the microwave field is applied adiabatically,
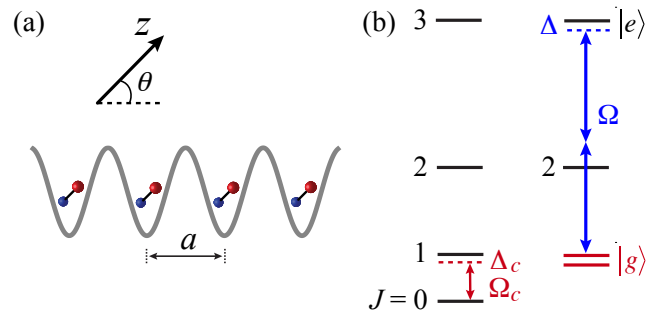

FIG. 1. (a) 1D array of polar molecules on an optical lattice tilted by the angle θ with respect to the laboratory z-axis. (b) Lowest rotational states of a polar $^1\Sigma$ molecule. A strong microwave field Ω_c couples the states $|0, 0\rangle$ and $|1, 0\rangle$, providing them with permanent dipole moments in the rotating frame; a weak field $\Omega \ll \Omega_c$ drives the two-photon transition between one of the dressed states $|g\rangle$ and the state $|e\rangle = |3, 0\rangle$, with the single-photon frequency far detuned from the $J = 2$ state. All fields are linearly polarized along z.

the molecules must all populate the same field-dressed state. We choose this state, $|g\rangle = a|0,0\rangle + b|1,0\rangle$, as the ground, 'spin-down', state. In the rotating frame, the state $|g\rangle$ has a permanent dipole moment $d_g = (\sqrt{2}ab/\sqrt{3})d$, leading to the dipole-dipole interaction between the molecules, $V_{\rm dd}(r) = d_g^2/r^3(1 - 3\cos^2\theta)$, whose magnitude and sign are tunable by changing Δ_c/Ω_c and θ [13]. We assume the coupling field to satisfy the condition $V_{\rm dd}(a) \ll \Omega_c \ll 2B$ that ensures that the dipole-dipole interaction does not mix $|g\rangle$ with the neighboring field-dressed state nor with the dark $|1,1\rangle$ state, which for typical experimentally realizible systems (molecules with dipole moment $1-5$ Debye and $a \sim 250 - 500$ nm) requires $\Omega_c \sim 1-100$ MHz. As the effective spin-up state we choose a level without a permanent dipole moment, $|e\rangle = |3,0\rangle$. The two pseudo-spin states, $|g\rangle$ and $|e\rangle$, are connected by a weak two-photon transition with Rabi frequency $\Omega \ll \Omega_c$, with the single photon resonance far detuned from the $|2,0\rangle$ state, which remains unaffected by the microwave field.

Using the rotating wave approximation, we can write the Hamiltonian for an ensemble of molecules on an optical lattice as

$$H = -\hbar\Delta \sum_i |e_i\rangle\langle e_i| + \frac{\hbar\Omega}{2}\sum_i(|e_i\rangle\langle g_i| + |g_i\rangle\langle e_i|)$$
$$+ V\sum_{j<i}\frac{|g_i,g_j\rangle\langle g_i,g_j|}{|i-j|^3}, \quad (1)$$

where $V = V_{\rm dd}(a)$ and $\Delta = \omega - \omega_{ge}$ is the detuning of the two-photon field from the $g - e$ resonance. Eq. (1) can be expressed via spin-1/2 operators S_α as

$$H = h_z\sum_i S_z^{(i)} + h_x\sum_i S_x^{(i)} + V\sum_{j<i}\frac{S_z^{(i)}S_z^{(j)}}{|i-j|^3}, \quad (2)$$

where $h_x = \hbar\Omega$, $h_z = -(\hbar\Delta + \zeta(3)V)$, and $\zeta(3) = \sum_{k=1}^\infty 1/k^3 \approx 1.202$ is Riemann's zeta function [14]. The magnitude and sign of the parameters h_z/h_x and V/h_x can be tuned by changing Δ, θ, and Δ_c/Ω_c. Note that the two-photon transition driven by Ω is key to realizing the dipole blockade [15] with molecular rotational levels: the pseudo-spin states $|g\rangle$ and $|e\rangle$ have a difference in angular momentum of $\Delta J \geq 2$, and therefore are not mixed by the dipole-dipole interaction. This leads to strong interactions between molecules in the $|g\rangle$ state, while eliminating the interactions between molecules in the $|g\rangle$ and $|e\rangle$ states (the "flip-flop" terms).

The thermodynamic properties of the Hamiltonian (2) have been studied before in the context of Rydberg atoms [16–18], and similar implementations based on polar molecules have also been discussed previously [19]. The main advantage of using polar molecules is that the dynamics occurs within the manifold of low-energy rotational states, whose lifetimes (> 1 s) are much longer
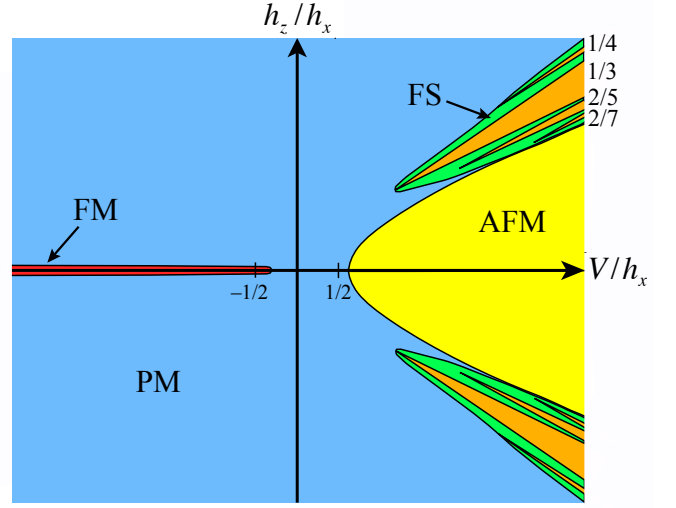

FIG. 2. Schematics of the ground-state phase diagram corresponding to the Hamiltonian (2). The commensurate crystal phases with indicated fillings (orange) are surrounded by the floating solid (FS) phase (green).

than the lifetimes of Rydberg states ($\sim \mu$s). Therefore, polar molecules offer the possibility of creating ordered structures with much larger size.

The main features of the phase diagram of the system described by Eq. (2) are shown in Fig. 2 and can be best understood in terms of the filling with which the minority spin component occurs. When V is positive and dominates over both h_z and h_x, the system is in an antiferromagnetic (AFM) phase, corresponding to half-filling, i.e. the up and down spins occur in an alternating order. In the absence of a longitudinal field, $h_z = 0$, with V being large and negative, all the spins are pointing in the same direction, which corresponds to the ferromagnetic (FM) ordering. There, any finite value of h_z breaks the $\mathbb{Z}_2$ symmetry of the model leading to the breakdown of the FM phase [20]. Note that the long-range interaction leads to the opposite shifts of the transition points on the $h_z = 0$ line. In the FM phase, the long-range terms lead to renormalization of the nearest neighbor coupling constant, while for the AFM interaction they result in frustration accelerating the melting of the phase. From both sides of the AFM phase there is a complete devil's staircase of crystalline configurations with different commensurate lattice spacings [17, 18, 21]. In general, the commensurate phases melt in two steps: first into the floating solid (FS) phase, and then into the paramagnet (PM). The FS phase is characterized by a finite density of Bose condensed dislocation defects of the commensurate crystal and exhibits a gapless excitation spectrum [17]. At the tip of the lobe at filling 1/3 and 1/4, however, the system belongs to the Potts universality class, which results in a direct transition from the commensurate phase to the PM [18].

Adiabatic preparation of the ordered phases, as e.g.

proposed in Ref. [19], is challenging due to the vanishing energy gap between phases on the phase boundary. For a system of N particles the gap closes as $1/N$ for the Ising transition between the PM and the AFM phases [22] and for the transition between the PM and the FS phases [23], and as $1/N^2$ for the transition between the FS phase and the commensurate crystals due to its relation to free fermions [5, 17]. Within the Landau-Zener approximation (equivalent to the Kibble-Zurek formalism for continuous phase transitions) for the adiabatic crossing of the phase boundary [22], the average size of the ordered domains is proportional to the square root of the sweep duration. While, in some cases, this scaling can be improved using non-linear sweeps [23, 24], it is highly desirable to have a more robust protocol. Here, we present a non-adiabatic method for creating crystalline order in the system, with the growth of the domains scaling much better than within the adiabatic approach.

In the following, we use the dipole blockade of microwave excitations to non-adiabatically construct strongly interacting ground state phases of the Ising model, eq. (2), as schematically shown in Fig. 3. We consider the case of $V > 0$, corresponding to $\theta = \pi/2$, and exemplify the technique by caclulations for ^{7}Li^{133}Cs molecules ($d = 5.520$ Debye, $B = 5.636$ GHz [25]) in an optical lattice with $a = 266$ nm.

The procedure to grow large ordered domains is schematically illustrated in Fig. 3. In the absence of the spin-flipping field Ω, all the molecules are initialized in the ground (spin-down) state $|g\rangle$. The energy needed to resonantly flip the first spin in the lattice is given by $E_1 = -2V\zeta(3)$, and we use it to renormalize the detuning as $\delta = \Delta - E_1$. In the first step, we nucleate the dipolar crystal using a π-pulse far detuned from E_1, which corresponds to $\delta/\Omega = \sqrt{N}$, where N is the number of sites of the ordered structure that one wishes to prepare. Then, each of the up spins separated by $\sim N$ sites serves as a center around which the ordered phase is to be grown.

Next, we make use of the dipole-dipole interactions between the molecules to deterministically grow the commensurate dipolar crystal with filling $1/n$ near the nucleation center. In order to resonantly flip a spin located n sites away from the nucleation center we use a π pulse with detuning $\delta = V/n^3$, while frequency flipping the third spin in the chain is shifted by $\delta = V/n^3 + V/(2n)^3 = 9V/(8n^3)$. Such detuned pulses will only affect these two spins, flipping any other spin in the system will be highly off-resonant. The size of the ordered structure that one can deterministically prepare with π pulses, Δx_0, is limited by the Rabi frequency Ω, which must be smaller than the change of the detuning δ from one pulse to another. Assuming $\Omega \sim 2\pi \times 50$ Hz and LiCs molecules, whose interaction is given by $V/\hbar = 2\pi \times 40.7$ KHz, one can deterministically prepare the states with $\Delta x_0 = 8a$ for the $n = 2$ (AFM) phase and $\Delta x_0 = 9a$ for the $n = 3$ phase.
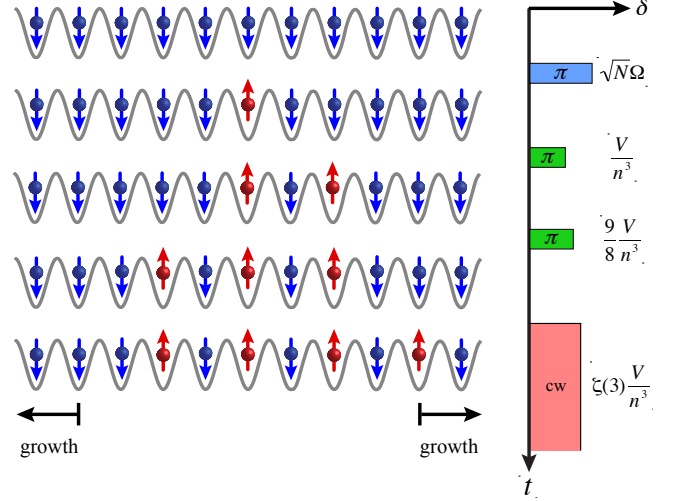

FIG. 3. Pulse sequence used to non-adiabatically prepare an ordered phase with a filling $1/n$: (i) a far detuned π pulse (blue) nucleates the phase by flipping one spin per N lattice sites; (ii) a few resonant π-pulses (green) deterministically create a few lattice cells of the phase near the nucleation center; (iii) continuous driving (red) propagates the crystal boundary. The required values for the renormalized detunings of the pulses, $\delta = \Delta - E_1$, are shown. The lattice plots exemplify the $n = 2$ (AFM) case.

After a few pulses the value of detuning required to flip the next spin in the ordered chain approaches the value of $\delta = \zeta(3)V/(na)^3$, and one can use continuous driving to grow the ordered structure further. Then, the dynamics of the crystal growth is equivalent to a single particle hopping on a semi-infinite two-dimensional lattice $\{i,j\}$ given by the domain size, $\Delta x_i \geq \Delta x_0$, and the domain center of mass position, $\bar{x}_i$, as given by the effective Hamiltonian,

$$H_{\text{eff}} = \frac{\hbar\Omega}{2} \sum_{i,j} \Big(|i,j\rangle\langle i+1, j\pm1| + |i,j\rangle\langle i-1, j\pm1| \Big)$$
$$+ V \sum_{\substack{i,j \\ k<i}} \frac{|i,j\rangle\langle i,j|}{(kn)^3}, \quad (3)$$

which is schematically shown in Fig. 4 (a). Numerically solving the time-dependent Schrödinger equation by exact diagonalization, we study the dynamics of the crystalline domain. As shown in Fig. 4 (b), the domain size increases linearly with time, illustrating that the growth process is highly efficient.

In order to prepare commensurate phases of higher order, such as 2/5 or 2/7, one can replace the continuous driving with a series of pulses, $\{\pi_1, \pi_2, \pi_1, \pi_2 \dots\}$, with detunings δ_1 and δ_2. In the case of the 2/5 phase, whose unit cell is given by $\uparrow\downarrow\downarrow\uparrow\downarrow$, the required detunings are $\delta_1 \approx 0.05\ V$ and $\delta_2 \approx 0.14\ V$. The frequency resolution needs to fulfill the condition, $\Omega < \delta_2 - \delta_1$, which can be
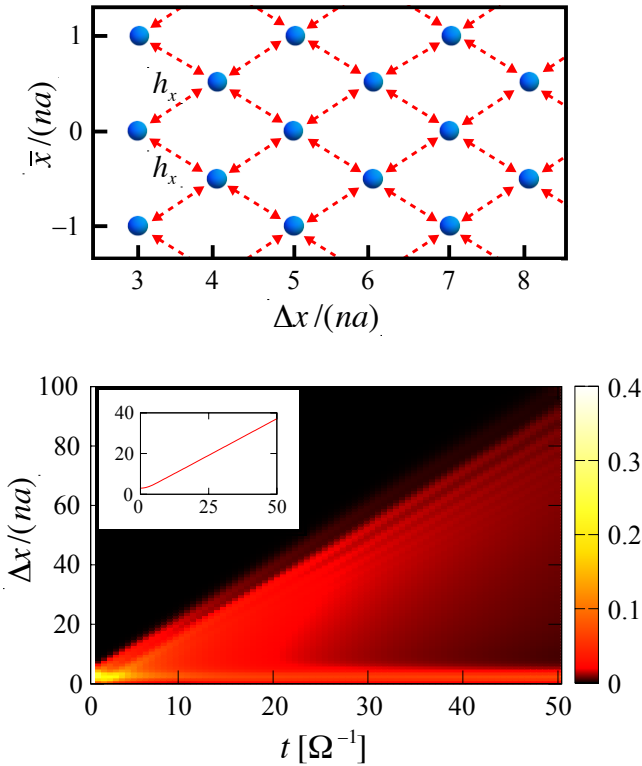
FIG. 4. Top: the growth of the crystalline phase can be visualized as a particle hopping on a 2D lattice, with Δx the domain size, and $\bar{x}$ the domain center-of-mass position, as given by the effective Hamiltonian, eq. (3). The example is given for $\Delta x_0 = 3na$. Bottom: the dynamics of the growth of the ordered phase during the continuous driving. Colors indicate the populations corresponding to different domain sizes, the inset (same axis labels as in the main figure) shows the root-mean-square of the size, which is linearly increasing with time.

easily satisfied for LiCs, requiring $\Omega \lesssim 4$ KHz.

The leading source of errors in the process of growing the dipolar crystal will be due to defects created by flipping a spin $n + 1$ sites (instead of n sites) away from the domain boundary. During the continuous driving, these states are detuned by $\Delta_{n+1} = V(1 + [\zeta(3) + \psi^{(2)}(1/n)/2]/n^3)$, where $\psi^{(n)}(z)$ is the polygamma function [26]. The number of spins that can be flipped before creating a defect can be estimated by the condition $N\Omega^2/\Delta_{n+1}^2 \approx 1$, which effectively limits the Rabi frequency. Since the number of spins one can flip within the lifetime τ of the system is given by $N \approx \Omega\tau$, we can calculate the average size of the domains to be $N = (\Delta_{n+1}\tau)^{2/3}$, which scales more favorably with time than the limit set by the Kibble-Zurek prediction. Assuming $V/\hbar = 2\pi \times 41\,\mathrm{kHz}$ and the lifetime of the system of $\tau = 1\,\mathrm{s}$, we find that it is possible to prepare domains of a $n = 2$ crystal consisting of up to $nN = 1700$ spins, while for the $n = 3$ crystal one can reach the size of $nN = 1000$ spins. The value of N determines the correlation length

of the defects, i.e. the effective temperature of the system, $T_{\mathrm{eff}} \sim \Delta_{n+1}/N$. Here, we find $T_{\mathrm{eff}} = 200$ pK for LiCs molecules and $N = 1000$.

We note that in principle the performance of this procedure can be improved even further, by using a composite pulse sequence that dynamically decouples the processes leading to the creation of defects [27]. Likewise, employing optical superlattice techniques [28] could also be used to improve the addressing of individual transitions.

Finally, we would like to emphasize that the dipolar crystals prepared after switching off the microwave driving correspond to the ground state of the Hamiltonian (2) in the limit of $h_x = 0$, and it is possible to explore the region of finite h_x by adiabatically turning the microwave driving field on again. Thus, it becomes possible to explore static and dynamic properties of the commensurate phases. In particular, the dynamics of fractionalized low-energy excitations can be studied, which are eventually responsible for the transition to the floating solid phase [17].

In summary, we have proposed a novel method to create strongly interacting many-body states in the dipole blockade regime of microwave transitions between rotational states of polar molecules. Our approach relies on the non-adiabatic driving of the microwave transitions and allows for the possibility to construct larger domains of dipolar crystals than in an adiabatic scenario. The scheme presented here is general and can be implemented with any two-state system possessing long-range Ising interactions, such as different spin states of $^2\Sigma$ or $^2\Pi$ molecules, trapped ions [29], Rydberg atoms, or Nitrogen-Vacancy centers in diamond [23]. Furthermore, our proposed method is not limited to dipolar crystals and could be applied to other systems where the initial state can be efficiently coupled to the many-body state of interest by non-adiabatic driving.

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